Light Emission Statistics as a Local Probe for Structural Phase Switching

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Abstract - The sensitivity of the spontaneous emission rate of an excited dipolar emitter to the local environment makes single-molecule spectroscopy a unique tool to sense optical and structural properties in its surrounding on the nanoscale. Modification of the emission rate has been reported in literature using well-defined structures, such as metal surfaces [1] or photonic crystals [2]. The increasing interest in the statistical properties of the spontaneous emission rates in complex media [3-6] is justified by their importance for many applications, such as molecular imaging or solar cells.
From a fundamental point of view, the emission rate is proportional to the number of available optical modes at the position of the emitter, i.e. proportional to the electromagnetic local density of states (LDOS)[7]. In random uncorrelated media, the LDOS fluctuations can be explained to some extent by a single scattering statistical model and are dominated by the near-field interaction with the nearest scatterer at the scale of the excluded volume [4]. Temporal lifetime fluctuations can then be correlated to fluctuations in the position of the nearest scatterer and provide a suitable probe for the dynamics of the structure around the emitter. The observed variance of experimental spatial fluctuations of LDOS in random photonic media and lifetime measurements in dense colloidal suspensions of weak scattering particles seem to be consistent with this single-scattering regime. However, the experimental distributions of emission rates in disordered highly scattering dielectrics can present disparate results ranging from non-Gaussian long-tailed statistics with very large decay rates [5], to nearly Gaussian distributions [6]. It can be argued that the differences in the experimentally retrieved decay rate distributions are attributed to effect of multiple scattering between nearby scatterers or long-range spatial correlations between scatterers.
In this work we show that the statistics of emission rates in correlated disordered media is extremely sensitive to the details of the radial distribution function around the emitter. We analyze the emission statistics for single emitter embedded in a finite cluster of resonant particles. However, instead of generate random configurations of scatterers, we compute the emission rates as the system evolves with time under equilibrium conditions. Assuming a standard Lennard-Jones (L-J) interaction between particles, this system is known to present a peculiar solid-liquid-like phase transition at finite temperature: Due to finite-size effects, the two phases cannot coexist at the melting temperature and the whole cluster presents an interesting dynamical behavior, switching between an amorphous solid-like phase and liquid-like phases [8]. This makes it an ideal model system to analyze the effects of local order on the emission rates. In the solid phase at low temperatures, the equilibrium positions are close to those corresponding to a face-centred-cubic (FCC) lattice, and the spectrum of emission rates present a strong chromatic dispersion reminiscent of the band structure of an infinite crystal of resonant dipoles, including spectral windows where the emission is enhanced and pseudo-gaps where it is dramatically inhibited. At the melting temperature, the total scattering cross section of the system does not present significant differences between the two phases while the emission rate jumps following the dynamics of the system. While light scattering measurements would be blind to such dynamical changes, the lifetime statistics would then provide a direct signature of a phase switching behavior.

References

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